

LA-UR-14-28883 (Accepted Manuscript)

Why is weapons grade plutonium more hazardous to work with than highly enriched uranium?

Cournoyer, Michael Edward Costigan, Stephen Andrew Schake, Bradley Scott

Provided by the author(s) and the Los Alamos National Laboratory (2016-08-15).

To be published in: Journal of Chemical Health and Safety

DOI to publisher's version: 10.1016/j.jchas.2014.10.004

Permalink to record: http://permalink.lanl.gov/object/view?what=info:lanl-repo/lareport/LA-UR-14-28883

Disclaimer

Approved for public release. Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the Los Alamos National Security, LLC for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396. Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; as an institution, however, the Laboratory does not endorse the viewpoint of a publication or guarantee its technical correctness.



Why Is Weapons Grade Plutonium More Hazardous to Work with than Highly Enriched Uranium?

Michael E. Cournoyer, Stephen A. Costigan, and Bradley S. Schake

Los Alamos National Laboratory Los Alamos, NM 87545 (505) 665-7616 mec@lanl.gov

ABSTRACT

Highly Enriched Uranium (HEU) and Weapons grade plutonium (WG Pu) have assumed positions of dominant importance among the actinide elements because of their successful uses as explosive ingredients in nuclear weapons and the place they hold as key materials in the development of industrial use of nuclear power. While most chemists are familiar with the practical interest concerning HEU and WG Pu, fewer know the subtleties among their hazards. In this study, a primer is provided regarding the hazards associated with working with HEU and WG Pu metals and oxides. The care that must be taken to safely handle these materials is emphasized and the extent of the hazards is described. The controls needed to work with HEU and WG Pu metals and oxides are differentiated. Given the choice, one would rather work with HEU metal and oxides than WG Pu metal and oxides.

Introduction

Highly enriched uranium (HEU) and Weapons Grade plutonium (WG Pu) have assumed positions of dominant importance among the actinide elements because of their successful uses as explosive ingredients in nuclear weapons and the place they hold as key materials in the development of industrial use of nuclear power. Their practical interest depends on their nuclear property of being readily fissionable with neutrons and their availability in quantity.

Uranium metal is heavy, silvery-white, malleable, ductile, and softer than steel. See Figure 1.



Figure 1. Uranium Metal before Its Surface Oxidizes to a Hard, Black Surface

It tarnishes in air, with the oxide film preventing further oxidation at room temperature.

After machining, the surface oxidizes, typically within hours, to a hard, black surface.

HEU used for nuclear weapons typically contains 93% (by weight) Uranium-235 (²³⁵U).² Uranium oxides are most commonly found in the forms of triuranium octoxide (U₃O₈) and uranium dioxide (UO₂). Both oxide forms are solids that have low solubility in water and are relatively stable (not "time sensitive") over a wide range of environmental conditions. U₃O₈ is the most stable form of uranium and is the form most commonly found in nature. UO₂ is the form in which uranium is most commonly used as a nuclear reactor fuel. At ambient temperatures, UO₂ will gradually convert to U₃O₈. Because of their stability, uranium oxides are generally considered the preferred chemical form of HEU for storage or disposal.

Plutonium metal has a bright, silver-like appearance at first, but it oxidizes very quickly to a dull gray. See Figure 2.



Figure 2. Plutonium Metal after Its Surface Oxidizes to a Dull Gray Surface

It is about as hard and brittle as gray cast iron unless it is alloyed with other metals to make it soft and ductile. Plutonium (IV) oxide is the chemical compound with the formula PuO₂. It can vary in color from yellow to olive green, depending on the particle size, temperature and method of production. Plutonium dioxide (PuO₂) is a stable ceramic material with an extremely low solubility in water; it is much less soluble than uranium. Due to the radioactive alpha decay of plutonium, PuO₂ is warm to the touch. WG Pu used for nuclear weapons typically contains in excess of 90% Plutonium-239 (²³⁹Pu).³

While most chemists are familiar with the practical interest concerning HEU and WG Pu, fewer know the subtleties among their hazards. In this study, a primer is provided regarding the hazards associated with working with HEU and WG Pu metals and oxides. The hazards of the many other forms of uranium and plutonium are beyond the scope of this primer. The major hazards associated with these materials are identified. The types of controls needed to work with HEU and WG Pu metals and oxides are discussed. Additional analysis of hazards and controls should be performed by qualified personnel prior to working with these materials based on the specific materials and operations under evaluation.

Definitions

• **Alpha Radiation** Particles with relatively large mass (consisting of two neutrons and two protons, i.e., He nucleus) and charge that travel only a few centimeters in air.

- Alpha particles are completely absorbed by thin materials such as a sheet of paper and by the layer of dead cells of human skin.
- **Beta Radiation** Particles with relatively small mass (i.e., an electron) and charge that travel centimeters to tens of meters in air depending on their energies. Most beta particles are capable of penetrating the skin and the lens of the eye.
- Gamma/x-ray Radiation Photons are generally highly penetration and their energy can be absorbed by the body's internal organs penetrate all the way through the body. Their ability to penetrate is roughly proportional to their energy. Gamma rays and x-rays are both photons but differ in their origin. Gamma rays originate from within the nucleus of an atom while x-rays originate from the electrons outside the nucleus.
- Neutron Radiation Particles with relatively moderate mass and no charge that are highly penetrating.
- by measuring the temperature at which a material spontaneously ignites in air. In a typical experiment, a sample is heated at a constant rate under flowing air while its temperature is continuously measured. The ignition temperature is the point at which deviation of the sample temperature from the programmed value indicates on-set of a self-sustained reaction.

Properties Associate with HEU and WG Pu Metals and Oxides

- **Reactivity** If uranium metal is left exposed to air oxidation of the surface of uranium occurs within a few hours, forming a hard black coating.¹ Plutonium, however, is much more chemically reactive than uranium. Under the same conditions, plutonium loses its metallic sheen quite rapidly, before it forms an olive green layer (fuzz-like appearance) of PuO₂ around the piece of metal.
- Pyrophoricity When tested under the same conditions, bulk samples of uranium must exceed 700°C before self-ignition occurs, while plutonium must exceed 500°C before self-ignition occurs.⁵
- **Toxicity** Based on the classifications published in the Official Journal of the European Communities, ⁶ ²³⁵U is in the Low Radiotoxicity classification (Group 4), whereas, ²³⁹Pu is in the Very High Radiotoxicity classification (Group 1). To be chemically toxic, HEU and WG Pu must be in their soluble forms. As HEU and WG Pu metals and oxides are insoluble, they are not chemically toxic.
- Criticality Anywhere from 0 to 7 neutrons may result from one fission; average for 235 U is ≈ 2.5 and average for 239 Pu is $\approx 3.0.^{8}$ The critical mass of a fissile material is the amount needed to sustain a nuclear chain reaction. The minimum critical mass of bulk HEU metal is about 50 kg, while the critical mass of bulk WG Pu metal is about 15.5 kg. Based on variables, especially physical dimensions (shape) and presence of neutron moderating materials such as water or other hydrogenous substances, the minimum critical mass can be significantly lower for both materials than those for bulk metal.

HEU and WG Pu Metals and Oxides Hazards

Storage Hazards The long-term storage of uranium ingots can form a pyrophoric surface caused by reaction with air and moisture. This is due the formation of powdered hydride and hydrogen. Moist dust, turnings, and chips react slowly with water to form hydrogen. These compounds in the presences of air represent fire and explosive hazards. Thousands of spontaneous fires have been experienced at room temperature during drum storage of lathe turnings or uranium briquettes made from compacted turnings. Over pressurization of storage drums resulting in release of radioactive material have been observed when fuel or scrap is found to be severely corroding in their containers. In many reported incidents, flashing of the fuel or explosions occurred when the containers were opened. The flashing was believed to be the spontaneous ignition of uranium or uranium hydride powder which became suspended due to the mechanical disturbance of opening the container.

These hazards are more severe with plutonium. In addition, plutonium expands up to 70% in volume as it oxidizes and thus may burst its container. Case studies show that mechanical wedging resulting from this expansion can even breach a second metal container, resulting in localized contamination release and possible exposure of personnel.

Long-term storage packaging needs are very different for HEU and WG Pu due to significant differences in reactivity. Most uranium samples require only one containment package, while plutonium samples require an inner convenience container and a vented (i.e., filtered vent) outer containment package. To prevent

oxidation, plutonium is stored in sealed cans or other sealed containers. Pressure can build up in the containers from both chemical and physical processes. Alpha particles from the radioactive decay of plutonium eventually become helium gas, which is usually harmless but can create enough pressure to burst a non-vented container. Containers should be monitored for bulging and repacked before pressure builds up and the containers burst.

Oxidation of the plutonium metal and rupture of the container by mechanical wedging are prevented if the storage container is hermetically sealed. Plutonium sesquioxide (Pu₂O₃) should be converted to PuO₂ before storage. Primary and secondary containers should be hermetically sealed and contain no plastics or other materials that decompose as a result of radiation exposure.¹²

Flammability Hazard Uranium in large block form does not present a significant fire risk. ¹³ Uranium in finely divided form is readily ignitable, and uranium scrap from machining operations is subject to spontaneous ignition. Grinding dust has been known to ignite even under water, and fires have occurred spontaneously in drums of coarser scrap after prolonged exposure to moist air. Because of uranium's thermal conductivity, larger pieces generally have to be heated entirely to their ignition temperature before igniting.

As with storage hazards, flammability hazards are more severe with plutonium. When heated to its ignition temperature, plutonium, even in large block, reacts at an accelerated oxidation rate, which sustains continued oxidation.

The spontaneous ignition of uranium can usually be avoided by storage under dry (without moisture) oil.¹³ Plutonium stored under oil has been reported to

spontaneous ignite.¹⁴ Plutonium should be stored as pure metal (Pu) or in its PuO₂ form in a dry, inert or slightly oxidizing atmosphere. Since fires involving uranium and plutonium burn relatively quickly through fines and turnings, limiting the amount of turnings throughout the machining area is important so that a small, localized fire does not propagate to other turnings or combustible materials.

The hazards involved in the control or complete extinguishing of metal fires include extremely high temperatures, steam explosions, hydrogen explosions, toxic products of combustion, explosive reaction with some common extinguishing agents, breakdowns of some extinguishing agents with the liberation of combustible gases or toxic products of combustion, and, in the case of HEU and WG Pu, dangerous radioactivity. Some agents displace oxygen, especially in confined spaces. Therefore, extinguishing agents and methods for their specific application must be selected with care. Metal fires should not be approached without suitable self-contained breathing apparatus and protective clothing, unless the fire is enclosed in a glovebox.

For plutonium metal, complete exclusion of oxygen and/or rapid heat removal are the effective ways to extinguish fires. Sand can also be used to smother and plutonium fires. Magnesium oxide sand is probably the most effective material for extinguishing a plutonium fire. Magnesium oxide cools the burning material, acting as a heat sink, and also blocks off oxygen. Because of reactivity and criticality concerns, water may not be the appropriate extinguishing agent.

• Internal Radiological Dose Hazard The primary routes of internal exposure from working with HEU and WG Pu metals and oxides are inhalation (breathing), ingestion (eating, swallowing) or injection (wounds) or absorption through damaged

skin. In most assessments, inhalation is considered the greatest hazard. Injection (i.e., a contaminated wound), although less likely to occur than inhalation, can result in higher internal dose than through the inhalation and ingestion pathways. Inhalation is the most likely route accidental uptakes of significant quantities of plutonium.

Ingestion without inhalation is considered much less likely to occur and is generally of less risk than inhalation since these compounds are relatively insoluble and the majority of the materials pass through the gastrointestinal system without absorption.

Inhaled airborne particles of insoluble uranium and plutonium compounds like HEU and WG Pu metals and oxides are generally deposited throughout the respiratory tract. Typically only the particles that make it to the deep lung tissue, the alveoli, are capable of being absorbed into the body via the bloodstream. Materials enter the blood through the alveoli in the lung do so at a rate proportional to their solubility. The process of transfer from the lung to the bloodstream may take years with insoluble uranium and plutonium oxides. Once in the bloodstream, distribution among the organs of the body and the excretion from the body are determined by their chemical properties and biological characteristics of the person exposed.

When comparing internal radiation dose hazard, HEU and WG Pu metals and oxides have the following similarities:

- o The radiation source is inside the body
- o Exposure continues once deposited in the body
- o Alpha-emitting radionuclides give highest dose
- o Relatively long half-lives, but ²³⁵U (7.04E+08 years) has a much longer half-life than ²³⁹Pu (24,000 years.)

o Respirable particles of greatest concern are generally considered to be 0.1 - 10 um (not visible)

On the other hand, HEU and WG Pu have significant differences in *Specific Activity* (amount of radioactivity in a certain mass of material) and intrinsic radiological hazard (the internal radiation dose resulting from intake of a given amount of radioactivity). WG Pu contains much more radioactivity than an equivalent weight of HEU. See Table 1.

Table 1. Specific Activity HEU and WG Pu Comparison.

	Highly Enriched Uranium	Weapons Grade Pu
Half-life (years)	7M (²³⁵ U) 245,000 (²³⁴ U)	24,000 (²³⁹ Pu)
Specific Alpha Activity (Ci alpha/kg)*	0.065	92.9

^{*}These values are for comparison purposes. Actual specific activity will depend upon a number of variables including the percent enrichment and concentration of other alpha emitting isotopes present.

The Specific Activity (alpha) ratio for WG Pu versus HEU is 1400:1.

When one compares the inhalation hazards of HEU and Pu, the inhalation radiation risks from Pu are much greater. See Table 2.

Table 2. HEU and WG Pu Radiation Dose Comparison.

	Radiation Dose per μCi Inhaled, rem (Sv) CED*	Radiation Dose per µg Inhaled rem (Sv) CED*	
HEU (93%)	24 (0.24)	0.0016 (0.000016)	
Weapons Grade Pu (new)	85 (0.85)	7.9 (0.079)	

*CED = committed effective dose based on ICRP 119 values. Doses shown are for comparison purposes. Actual doses would depend upon a number of different variables that include material composition, chemical form and other factors.

CED is an acronym for committed effective dose which is a predicted dose to the individual resulting from a radionuclide intake. In the United States, the current regulatory annual dose limit for radiological workers is 5 rem (0.05 Sv). 16 Table 2 illustrates that when comparing HEU and WG Pu on a mass basis (µg inhaled), the radiation dose from WG Pu can be nearly 5000 times greater than that for HEU. Indeed, inhalation of only 1 µg of WG Pu could result in a dose exceeding the regulatory dose limit for workers.

Controls include confinement in ventilated hoods. Ventilated hoods are used to prevent airborne contamination. A hood, or slotbox, is not completely sealed; it has an opening or slot to allow you to reach in. Thus, a hood is an example of "confinement" because it has an opening through which you have direct access to the material. Working areas should be classified according to the relative radiotoxicity of the radionuclides; taking into account the nature of the operations and the total amount of material used. For example, an open-front hood is a confinement device designed to confine through air movement and to exhaust radioactive materials in

gaseous, vapor, or particulate form. An example of safety limits for use in open-front hoods for HEU and WG Pu in a nuclear facility at Los Alamos National Laboratory are listed in Table 3.¹⁷

Table 3. HEU and WG Pu Open-Front Hood Limit Comparison.

	Radionuclide		
Operation/ Material Type	HEU	WG Pu	
Storage and Stock Solution (g)	40000	60.0	
Simple Wet Operations (g)	4000	6.0	
Normal Operations (g)	40	0.6	
Open Powders (g)	4	Not permitted	

Normal Operations are considered work with bulk solids, metals, and other non-dispersible forms. Operations involving powders or easily dispersible material of WG Pu are not allowed in open-front hoods without supplemental analysis, authorization and compensatory measures. Forty grams and 4 grams of HEU metal and oxide can be processed in an open-front hood, respectively. On the other hand, only 0.6 gm of WG Pu metal and no amount WG Pu oxide can be processed in an open-front hood.

Because plutonium is of particular concern if inhaled, controls (including containment in sealed gloveboxes) are used to prevent airborne contamination. A glovebox is illustrated in Figure 3.



Figure 3. The Inside of Glovebox Used to Process Plutonium

A glovebox has ports with long-sleeved gloves attached that allow material to be sealed in the glovebox. A glovebox is an example of "containment" because it is completely sealed with no openings. During operations such as glove changes or bagouts that breach a containment system, respirators are used to minimize the chance of inhaling plutonium.¹⁸

Ingested insoluble compounds are poorly absorbed by the gastrointestinal tract and are retained in the body for only a short time, therefore generally having a low toxicity, as compared to soluble compounds. HEU and WG Pu metals and oxides are insoluble compounds. Most inhaled particles are deposited above the alveolar region of the lung and are moved by ciliary action back up the trachea and then transferred to

the gastrointestinal tract by swallowing. Since most of the inhaled and ingested uranium is not absorbed it leaves the body in the feces. Most of the Uranium that is absorbed by the lungs is deposited primarily in the kidneys, bones, and liver. Absorbed Plutonium is primarily absorbed by bone surfaces and the liver. Uranium and Plutonium that is absorbed is eventually excreted from the body in the urine. The rate of excretion from the body differs greatly between HEU and WG Pu being approximately 12% *per day* for HEU and 1 - 2% *per year* for WG Pu. If plutonium were to enter a worker's lungs today, much of the plutonium would still be in his body 30–50 years later.

Intact skin is a good barrier against most forms of uranium and plutonium including HEU and WG Pu metals and oxides, so absorption through skin is extremely rare. Absorption through wounds, though less common than inhalation, can be extremely hazardous.²¹ Large amounts of radioactive or toxic material could be deposited directly into the body through injection and then absorbed into the bloodstream. Absorption through wounds typically results from accidents with contaminated sharp objects.²²

Excision is the surgical removal of contaminated tissue. If a large amount of contamination is located at the wound site, excision can dramatically reduce the exposure. Dose reductions of up to a factor of 100 have been achieved with excision.²³ Usually only a small amount of tissue is removed, which does not present a significant health hazard.

All potential intakes of HEU and WG Pu should be reported to and evaluated by qualified Health Physics professionals to determine the need for further analysis and possible treatment by Medical professionals.

- External Radiological Dose Hazard When comparing external radiation dose hazard, HEU and WG Pu have the following similarities:
 - Source of radiation outside the body
 - o Capable of penetrating container, shielding, clothing, skin
 - o X-rays, gamma and neutron radiation
 - o Radiation dose stops when source removed

Most external radiation associated with uranium comes from beta particles emitted by one of uranium's radioactive decay products, Protactinium-234m (234mPa). These high-energy betas, ~2.3 MeV can travel about 8 meters in air. Beta particles cause a shallow dose—a radiation dose that penetrates the skin about 0.07 mm. Therefore, a person working several meters from this source could receive beta exposure to the skin or lens of the eye. Handling uranium can result in elevated dose rates to the skin of several hundred mrem/hour.

When comparing external dose hazard, external radiation is generally more of a concern for WG Pu than for HEU. Most gamma and x-ray radiation from plutonium is low energy and moderately penetrating. More penetrating (60 keV) gammas are emitted by Americium-241 (²⁴¹Am), which "grows in" as Plutonium-241 (²⁴¹Pu), which is almost always present in plutonium compounds, decays. In WG Pu that is more than 10 years old (since purification), these gamma rays are usually the source

of most of the external radiation.²⁴ Thus, the external radiation from plutonium increases over time from the decay of ²⁴¹Pu.

External and extremity dose is controlled by minimizing time, maximizing distance, using shielding, and using source reduction, collectively referred to as *as low as reasonably achievable* (ALARA) measures. Alpha particles emitted by uranium and plutonium are shielded by skin or paper and so present no external hazard. Beta radiation from uranium can be shielded with about 5 to 10 mm of plastic, rubber, or other material with a low atomic number. Shielding for beta, which is made of anything heavier than aluminum, produces bremsstrahlung ("braking") radiation. As the speeding beta particle approaches the positively charged nucleus of a large atom, it changes direction as it is attracted to the large, positively charged nucleus. This sudden change of direction causes the release of secondary radiation in the form of x-rays. This bremsstrahlung radiation produced is more penetrating and hazardous than the original beta radiation that is being shielded. Shielding material with a low atomic number (such as hydrogen found in water and plastics) should be placed next to beta sources to reduce production of these secondary x-rays.

Protective clothing may also be needed to reduce beta dose. Such protective clothing may include glass or plastic safety glasses and heavy rubber or leather gloves to reduce the beta dose to one's hands. Unusually heavy gloves eliminate the beta radiation dose to one's hands. Such gloves might interfere with one's dexterity while completing a task involving a beta radiation source.²⁵

Even when beta particles are shielded, significant penetrating gamma radiation may remain an exposure hazard. The most penetrating high-energy gamma

rays, up to about 5 mrem per hour, come from the uranium decay products. One can reduce the high-energy gamma radiation from these uranium decay products by using lead (usually 5 cm or more thick) or concrete (usually greater than 20 cm thick) shielding after shielding for the beta particles.

With freshly purified plutonium, most of the radiation comes from "soft" (~17 keV) x-rays, which penetrate several centimeters of soft tissue but are shielded by the steel walls of a glovebox or by lead aprons. These x-rays easily penetrate the rubber gloves in a glovebox, resulting in radiation dose to the hands. Gloves with a layer of lead can be used to reduce the extremity dose. Leaded-glass windows and lead storage containers further reduce dose from these sources.

Neutrons are more penetrating than gamma rays in the sense that they are typically more difficult to reduce through shielding Most of the neutron radiation emitted from HEU and WG Pu are the result of nuclear reactions (referred to as alpha-n reactions) where an emitted alpha particle interacts with the nucleus of a low atomic weight atom (e.g., Li, Be, N, O, F) present as impurities which causes the atom to emit a neutron. There is a strong dependence on the concentration of low-atomic mass elements in determining the neutron radiation hazard. The dose rate from neutron radiation emitted by enriched uranium can be up to 4 mrem per hour but for laboratory quantities is often not measurable or significant enough to warrant concern or special controls. Neutron emissions from WG Pu can be significantly higher than for HEU and may become the dominant radiation hazard when large quantities of WG Pu are present. The most effective shielding includes a neutron moderator such as several centimeters of water, oil, or plastic. Some gloveboxes have walls filled

with one of these substances and thick plastic windows. Typical shielding for neutrons is 30 cm or more of concrete or plastic. High-energy neutrons are usually shielded using a three-layer configuration:

- First layer—chosen for its high inelastic scattering (such as iron)—removes
 energy rapidly from neutrons, dropping them to a fast-neutron category.
- Second layer—containing a high hydrogen content material (water, wax, plastic, concrete)—moderates, slows, and thermalizes these fast neutrons.
- Third layer—possible materials include cadmium, boron, and lithium—reduces the flux of the resulting thermal neutrons to a reasonable level and reduces gamma photons created by the process of neutron absorption.

The effectiveness of this shielding should be checked by direct measurement. Health Physics professionals should always be contacted for evaluating radiation shielding before it is installed or modified.

Table 4 shows the approximate beta, gamma, and x-ray dose rates from 1-kg spheres (radius of 2.54 centimeter) of HEU and WG Pu.

Table 4. External Dose Hazard Comparison.

		Dose Rate (rem/hour)			
Grade	Isotope	Surface	Shielded	Shielded at 30 cm	Primary Radiation
Weapons	²³⁵ U	0.02	< 0.02	< 0.001	Beta Gamma
Weapons (New)	²³⁹ Pu	2	0.1	0.001	Gamma Neutron
Weapons (Old)	²³⁹ Pu, ²⁴¹ Am	3	1	0.01	Gamma Neutron

The table compares the "surface" dose rates to the hands when shielded only by thin latex gloves, the "shielded" dose rates when leaded gloves or steel containers are used, and the dose rates when shielded by the glovebox and a distance of 30 cm (that is, outside a glovebox). External hazards can be effectively reduced and managed for both with appropriate packaging or shielding.

With spheres, the surface dose rates of 1-g spheres are the same as for 1-kg spheres, but the 30-cm rates are 100 times less because the surface area is 100 times smaller. With flat sheets, the surface dose rates are also the same, but the 30-cm rates increase with the surface area. These rates are approximate; they depend on the composition, the shielding thickness, and the age in the case of WG Pu (which determines the amount of ²⁴¹Am), as discussed above.

• **Criticality Hazard** As previously discussed in this journal, a critical mass of HEU or WG Pu may become supercritical at which it emits lethal amounts of neutrons and gamma rays.²⁷ Neutron dose rates from criticality exposure are likely to be serious—if not fatal—to nearby personnel. Facilities with operations that could result in criticality accidents have criticality detection systems to warn workers of an accident.

A criticality accident may last for less than a second or may continue for hours. Many controls are in place to prevent a criticality accident, including the following:

- Maintaining a subcritical mass—the smaller the mass, the more likely a neutron will escape the material without colliding with a nucleus
- Maintaining separation—the spacing of subcritical units ensures that a critical mass does not accumulate
- Controlling the shape—the shorter the path a neutron has to travel though the
 material before it can escape, the less likely it will collide with a nucleus (longthin shapes are safest)
- o Controlling moderation—a subcritical mass could become critical in the presence of moderators, such as water, plastic, carbon and other light elements that are effective in slowing down neutrons
- Controlling reflection—neutron scattering in reflectors can return neutron leaking from a system for a second chance to cause fissions (almost any material can reflect neutrons)
- Using neutron poisons—poisons such as boron and cadmium capture neutrons

Discussion

Due to the difference in reactivity, uranium metal can be handled outside a glovebox, as shown in Figure 1, while plutonium metal can only be worked on in a dry, inert or slightly oxidizing atmosphere, as shown in Figure 2. While there have been many reports that uranium metal powder or chips and plutonium in these forms and in large

pieces will ignite spontaneously in air at ambient temperature, ¹⁰ uranium and plutonium metals are not pyrophoric at room temperature. ²⁸ The potential for uranium and plutonium fires is greatest when these metals are being cut, drilled, or machined. The small chips and shavings can readily ignite when heated by friction from machining tools. During storage, uranium and plutonium ingots can form a pyrophoric surface caused by reaction with air and moisture. This is due to hydride formation.

A variety of metals burn, particularly those in finely divided form.¹³ Some metals burn when heated to high temperatures by friction or exposure to external heat; others burn from contact with moisture or in reaction with other materials. Because accidental fires may occur during the transportation of these materials, it is important to understand the nature of the various fires and the hazards involved.

Pyrophoric ignition of plutonium is of particular concern because formation of plutonium oxide is an essential process for entrainment and dispersal of plutonium containing particles from a massive metal source. Limited amounts of water contributes to the intensity of a fire in uranium or plutonium, and greatly increase the contamination cleanup required after the fire. Smoke from fires involving radioactive materials can cause more property damage than the fire itself.

Alpha particles do not penetrate the dead layer of skin on the body. However, when they are in close contact with living cells inside the body, they are hazardous.

Although some external exposure is expected when working with uranium and plutonium, internal exposure is not. To preclude excursions of alpha emitters into the operator's breathing zone, ventilated hoods are used to confine uranium during laboratory

work with open powders, as shown in Table 3. Under the same conditions, gloveboxes are used to confine plutonium.

Uranium and plutonium are chemically toxic, as are other heavy metals such as arsenic and lead. While the chemical toxicity of uranium is comparable to uranium's radioactive toxicity, the chemical toxicity of plutonium is minor compared with plutonium's radioactive toxicity. If ingested, plutonium is considerably more hazardous to humans than uranium.

Externally penetrating radiation affects cells directly. External exposures may be fairly uniform over the whole body (external dose) or non-uniform, i.e., primarily focused on a limited body location (extremity dose). Excess external dose generates stochastic effects consisting of cancer and benign tumors in some organs. ¹⁶ Cancers induced by radiation do not have a threshold level of dose. Direct doses from radiation sources external to the body are measured by thermoluminescent dosimeters. Results from thermoluminescent dosimeters used by glovebox workers are received monthly. If glovebox workers' external or extremity dose approaches recommended limits, they are removed from the source. In other words, external or extremity dose can be controlled.

The consequences from an intake of uranium or plutonium are much more unpredictable. For example, the range of CED from the three similar puncture wound injuries with plutonium through a glovebox glove reported at LANL, since June of 2006, was 38 millrem to 7.5 rem.²⁹ There are numerous factors in play both prior and subsequent to an intake scenario that determine dosimetry result. Thus, there is great uncertainty in the internal dose outcome. Dose assessment from WG Pu intakes typically

requires collection of urine over weeks to months following a suspect intake. The dose assessment from WG Pu may require as much as six months to one year.

Systems that could become critical are carefully controlled. Personnel who work with significant amounts of fissile material greater than 500 g (or larger than a golf ball) must receive specific job and site training in criticality. Personnel who have not received criticality training must not touch or move fissile material.

Compared to all other actinides, uranium metal is rather well understood, mostly due to the fact that it is technologically relevant.^{30,31} On the other hand, plutonium metal has numerous layers of complexity, due to its unique position between localized and delocalized 5f states.³² While there are many criteria that relegate plutonium to the category of highest toxicity and greatest potential hazard, there have been no demonstrable injuries attributable to less than supra-critical concentrations of plutonium.³³

In summary, HEU metal can be worked on in a hood. WG Pu metal must be handled in a moisture-free (dry) or oxygen-free (inert) atmosphere in a glovebox. HEU metal can be stored under oil. WG Pu metal catches fire under the same conditions. Plutonium is three orders of magnitude more toxic than HEU. Criticality accidents are three times more likely with WG Pu than with HEU. The physical and chemical properties of HEU are predictable, while same properties for WG Pu are complex.

Conclusions

WG Pu can present up to 5000 times more radiological risk to workers than an equivalent mass of HEU. Increased knowledge of Pu physiological and toxicological behavior has kept pace with its greater availability. Given the choice, one would rather work with HEU metal and oxides than WG Pu metal and oxides.

Acknowledgements

The authors would like to acknowledge the U.S. Department of Energy and LANL's Plutonium Science & Manufacturing directorate for support of this work.

References

- 1. Morss, L.R., Edelstein, N., Fuger, J., (Eds.), The Chemistry of the Actinide and Transactinide Elements, 4rd ed. Springer: Dordrecht, The Netherlands, 2010.
- 2. http://www.wise-uranium.org/pdf/reenru.pdf, link verified September 15, 2015.
- Cournoyer, M.E.; Staudhammer, K., Los Alamos National Laboratory, Los Alamos, New Mexico, February 2005, LA-UR-04-0909.
- 4. Martz, J. C., et al., J. of Nuclear Materials **1994** 210:130.
- Hightower, J. R.; Trabalka, J. R., Argonne National Laboratory, Chicago, IL, February, 2000, ANL/TD/CP-84441.
- Shleien, B.; et al, Handbook of Health Physics and Radiological Health, 4rd Ed.
 Williams & Wilkins, 2012.

- Radiological Safety Training for Plutonium Facilities, August 2001, DOE-HDBK-1145-2001.
- 8. Mitchell, M.V., et al., Los Alamos National Laboratory, Los Alamos, New Mexico June 2014, LA-UR-14-24428.
- Monahan, S, "Criticality Safety and MOX," Actinide Research Quarterly, 1st/2nd
 Quarters, 2007.
- 10. Pearson, H. E., Hanford, Richland, WA, September 1954, HAN-64841.
- 11. Plys, M.G., Fauske & Associates, Inc., Burr Ridge, IL, April 2000, SNF-6192-FP.
- 12. Assessment of Plutonium Storage Issues at Department of Energy Facilities, January 1994, DOE/DP-123T.
- Primer on Spontaneous Heating and Pyrophoricity, December 1994, DOE-HDBK-1081-94.
- Haschke, J.M.; Martz, J.C., Los Alamos National Laboratory, Los Alamos, New Mexico, July 1993, LA-12624-MS.
- 15. Nenot, J. C.; Stather, J. W., The Toxicity of Plutonium, Americium and Curium, Elsevier, January, 1979.
- 16. 1990 Recommendations of the International Commission on Radiological Protection,
 Annals of the ICRP, 1990, 21(1-3), 12.
- 17. Cournoyer, M.E., et al., J. Chem. Health Saf. **2009** 16(1), 4-10.
- 18. Rael, D.G., et al., "Retrofit of an Engineered Gloveport to a Los Alamos National Laboratory's Plutonium Facility Glovebox," Journal of the American Society of Mechanical Engineers, Proceeding from WM'08, Phoenix, Arizona, February 24 - 28, 2008.

- 19. http://www.atsdr.cdc.gov/csem/csem.asp?csem=16&po=9, link verified September 15, 2015.
- 20. http://www.atsdr.cdc.gov/toxprofiles/tp143.pdf, link verified September 15, 2015.
- Hardy, M.W., et al., Los Alamos National Laboratory, Los Alamos, New Mexico February 2007, LA-UR 07-1305.
- 22. Balog, J.A., et al., Los Alamos National Laboratory, Los Alamos, New Mexico September 2008, LA-UR 08-06131.
- Radiological Safety Training for Plutonium Facilities, April 2008, DOE-HDBK-1145-2001.
- 24. Cournoyer, M.E., et al., "Replacement of Lead-Loaded Glovebox Glove with Attenuation Medium That Are Not RCRA-Hazardous Metals," Proceeding from the 17th Pacific Basin Nuclear Conference, Cancun, Mexico, October 25-29, 2010.
- 25. Castro, A.M., et al., J. Chem. Health Saf. **2012** 19(2), 3-10.
- Landsberger, S. et al., Los Alamos National Laboratory, Los Alamos, New Mexico June 2011, LA-UR 11-02620.
- 27. Cournoyer, M.E., et al., J. Chem. Health Saf. 2014, in press.
- 28. Totemeier, T.C., Argonne National Lab., Idaho Falls, ID, June 1995, ANL/ED/95 -2.
- 29. Cournoyer, M.E.; et al, J. Chem. Health Saf. **2011** 18(1), 22-30.
- 30. Lander, G. H., et al., Adv. in Phys. **1994** 43, 1.
- 31. Fisher, E. S., 1994, J. Alloy Comp. **1994** 213-214, 254.
- 32. Bradley, J.A., et al., Phys. Rev. B **2012** 85, 100102.
- 33. Wick, O.J. (Ed.), Plutonium Handbook A Guide the Technology, Vol. 1, American Nuclear Society, La Grange Park, IL, 1980.